ORIGINAL ARTICLE

Improvement of hybrid jig separation efficiency using wetting agents for the recycling of mixed-plastic wastes

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Abstract

We have developed the hybrid jig which combines the principles of jig separation and flotation. However, the selectivity of bubble attachment in water was poor because most plastic have inherently hydrophobic surfaces, so development of surface modification techniques for plastic particles would expand its application to the material recycling of plastics.

In this study, the hybrid jig separation of the polypropylene with glass fiber (PPGF) and high impact polystyrene (HIPS) having similar SG and surface wettability were investigated with three wetting agents (Di-2-ethylhexyl sodium sulfosuccinate (Aerosol OT, AOT), sodium lignin sulfonate (NaLS), and tannic acid (TA)). The results showed that the probability of bubble attachment was influenced by wetting agents because of their strong effects on the surface tension of solution and surface wettability of plastics. These results suggest that wetting agents could be utilized to control the selectivity of bubble attachment and improve the hybrid jig separation efficiency. In addition, since the hybrid jig separation of polyvinyl chloride (PVC) and polyamide, nylon-66 (PA) using AOT was imperfect, a two-step approach, composed of a pre-wetting step (first step) in a solution containing the wetting agent (AOT) and hybrid jig separation in water (second step), was proposed.

Keywords: Recycling, jig, hybrid jig, plastic, wetting agent

Introduction

The total global production of plastics has grown from around 1.5 million tons in 1950 to 348 million tons in 2017 and worldwide consumption has been increasing annually at a rate of 5% with the largest increase reported in Asia [1, 2]. There are two common ways to recycle plastics: (i) material recycling whereby plastic wastes are recovered for the production of new plastics, and (ii) thermal recycling whereby plastic wastes are used as fuel for power generation. Between the two, material recycling is more sustainable and profitable but requires exceptionally efficient separation of different types of plastics from mixed-plastic wastes to obtain products with very high purities (>99.9%). Achieving this very high plastic purity in recycling is very difficult, so mixed-plastic wastes are more commonly used in thermal recycling. In recent years, however, several studies have utilized mineral processing techniques to separate mixed-plastic wastes into their individual components to facilitate material recycling (e.g., gravity separation [3-10], dense medium separation [11-13], electrical separation [14-16], flotation [12, 17-28]).

Flotation is one of the common mineral processing techniques for fine fraction (minerals: -75 μm; coal: -150 μm) since normally, fine grinding is required (μm) for ores to achieve sufficient liberation of target minerals [9, 29]. Most of minerals have hydrophilic surface so, a collector (e.g., xanthate, aerofloats [30]) is usually added to selectively change the surface wettability to enhance the separation efficiency. In contrast, plastic flotation is usually carried out with wetting agents (e.g., AOT [22], NaLS [22], CaLS [12, 22-24], TA [22, 25, 26], PVA [22, 26-28]) since most plastic have inherently hydrophobic surfaces. However, for resources recycling, especially plastics, sufficient liberation is already achieved in even at coarser particle sizes (mm to cm) [30-32]. Moreover, additional size reduction (i.e., crushing and grinding) required for flotation that incurs higher costs and energy is needed [30].

One potentially effective technique for the separation of coarser size of plastics from mixed-plastic wastes is the use of jigs. Jigs can separate the particles at coarse size fractions (+0.5 mm) and jig is a gravity concentrator that separate the particles based on differences in their densities or specific gravities (SG) [7, 30, 33]. Among the various types of jigs, the TACUB (Takakuwa air chamber under bed) jig, commercially marketed as BATAC jig, is a popular and commonly used type of jig especially in coal cleaning because of its more uniform pressure distribution inside the separation chamber that provides higher separation efficiency. This uniform pressure distribution is facilitated by a series of pneumatically operated multiple air chambers, usually two cells under the separation chamber, that extend to its full width. Pressurized air is injected into the air chambers, causing water within the separation chamber to pulsate and induce stratification [3, 6, 30].

The TACUB jig was, however, ineffective for mixed-plastic wastes because SG differences of the various types of plastics in mixed-plastic wastes are very small. To address this problem, Tsunekawa et al. [3] developed the RETAC jig (R&E, Co., Ltd., Japan), an improved version of the TACUB jig, that facilitates plastic-plastic separation via precision control of the wave form during jig separation (Fig. 1(a)) [3, 4]. The RETAC jig, for example, successfully separated polystyrene (PS), acrylonitrile butadiene styrene (ABS), and polyethylene terephthalate (PET) from crushed copy machines [3]. Although effective, separation efficiency of the RETAC jig dramatically decreases when the density difference of plastics to be separated becomes small. Unfortunately, many types of plastics developed in recent years for identical applications have very similar SG. For example, two types of plastics used in the manufacture of large home appliances, polypropylene with glass fiber (PPGF) and high impact polystyrene (HIPS), both have SG equal to 1.04. In theory, separation of materials with similar SG is impossible using gravity separation techniques like jigs.

A new type of jig called the hybrid jig was developed to address this limitation. The hybrid jig works by combining the principles of gravity separation and flotation and could separate materials with similar SG so long as their surface wettability properties are different (Fig. 1(b)) [5]. Separation occurs due to the attachment of bubbles to more hydrophobic plastic particles that make them lighter than the less hydrophobic plastic particles that causes stratification during water pulsation.

In a previous study of the authors, hybrid jig separation of pure plastic mixtures (i.e., polyethylene (PE), polyethylene terephthalate (PET), polyvinyl chloride (PVC)) in water was carried out and good separation efficiency was reported [5]. However, the selectivity of bubble attachment in water for other plastics was poor because most plastic have inherently hydrophobic surfaces, so development of surface modification techniques for plastic particles would expand its application to the material recycling of plastics. This means that surface modification of plastics is required for the improvement of mixed-plastic waste separation using the hybrid jig, so in this study, the effects of wetting agents on bubble attachment probability and hybrid jig separation efficiency were investigated. In addition, a two-step approach to improve hybrid jig separation efficiency using wetting agents is proposed.

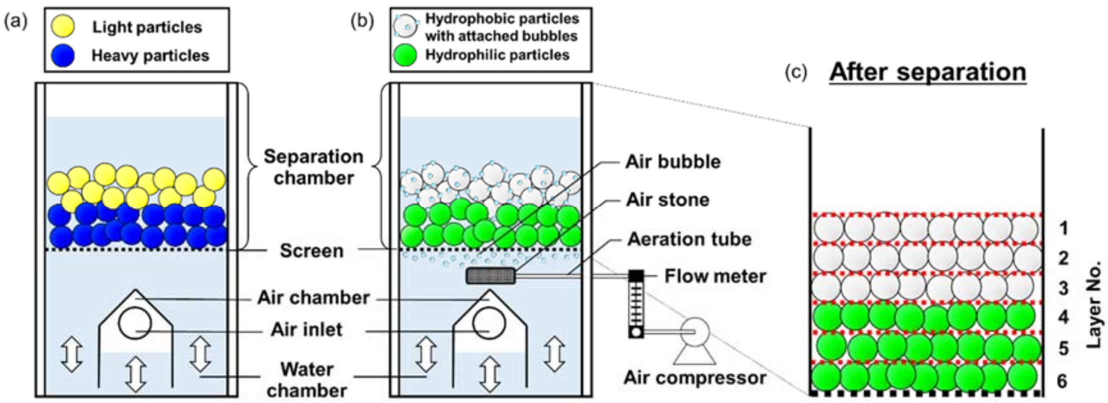

Fig. 1 Schematic illustrations of (a) RETAC jig, (b) Hybrid jig, and (c) how the products were partitioned into 6 layers after separation.

Materials

Samples

Polypropylene with glass fiber (PPGF) and high impact polystyrene (HIPS) obtained from a recycling facility of home appliances in Japan, and virgin plastic boards of polyvinyl chloride (PVC) and polyamide, Nylon-66 (PA) were used in this study and the SG of these samples are listed in Table 1. The virgin plastic boards were crushed by an orient mill (VH16, Seishin Enterprise Co. Ltd, Japan) and screened to obtain suitable size fractions for the hybrid jig separation experiments.

Table 1. Plastic samples and their specific gravities (SG).

Samples	Abbreviation	Specific gravity
Polypropylene with glass fiber	PPGF	1.04
High impact polystyrene	HIPS	
Polyvinyl chloride	PVC	1.38
Polyamide, Nylon-66	PA	1.37

Reagents

Methyl isobutyl carbinol (MIBC, Wako Pure Chemical Industries Ltd., Japan), a reagent widely utilized in flotation to stabilize bubble formation in solution, was used in the hybrid jig separation experiments. Three types of wetting agents were evaluated in this study: (1) Di-2-ethylhexyl sodium sulfosuccinate (Aerosol OT, AOT, Wako Pure Chemical Industries Ltd., Japan), (2) sodium lignin sulfonate (NaLS, Tokyo Chemical Industry Co. Ltd., Japan), and (3) tannic acid (TA, Wako Pure Chemical Industries, Ltd., Japan).

Experimental methods

Surface tension and contact angle measurements

Surface tension of solutions containing each of the wetting agents was measured using a temperature-controlled reaction vessel connected to a tensiometer (Krüss K100, Krüss GmbH, Germany). For the contact angle measurements, an air bubble generated from a syringe needle was introduced underneath the plastic sample submerged in solutions containing each of the wetting agents and the contact angle (angles between particle surface (solid-liquid interface) and bubble surface (gas-liquid interface)) (Fig. 2) was measured using a high-resolution digital microscope with image analysis capability (VHX-1000, Keyence Corporation, Japan).

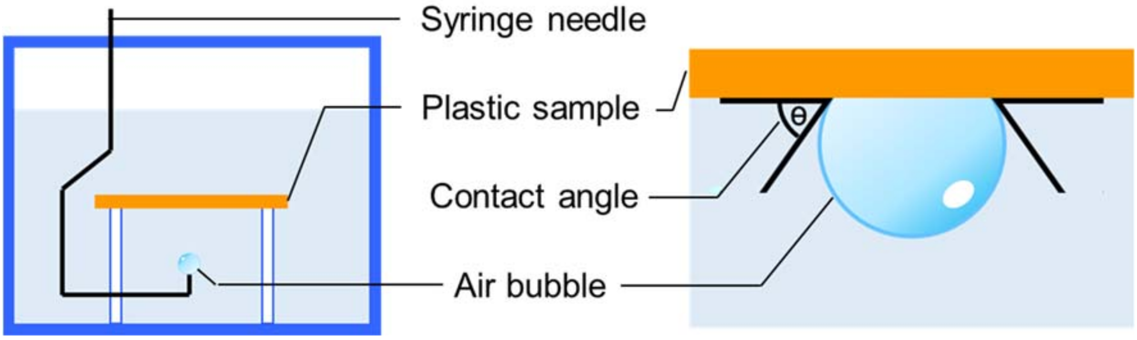

Fig. 2 Schematic diagrams of the contact angle measurement

Adsorption and desorption experiments

To obtain adsorption isotherm of AOT on PA [34], the adsorption experiments of AOT on PA were carried out. The particles of PA (25 g) were stirred (400 rpm) in to AOT solution (30, 50, 80, 100, 150, 180, 200, and 250 ppm) for 10 min then, left for 120 min. The dissolved sulfur concentration of remaining solution was measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES, ICPE-9820, Shimadzu Corporation, Japan) (margin of error = ±2%).

For desorption experiments, PA particles were put in 10 ppm AOT solution for 3 min and then, washed by water. The particles were then put in another beaker filled with water and the dissolved sulfur concentration of solution was measured by ICP-AES after 30, 60, 90, and 120 min to calculate the desorption amount of AOT from PA.

Hybrid jig separation experiments

Fig. 1(b) shows a schematic diagram of the desktop hybrid jig separation apparatus used in this study. This device is a modified desktop RETAC jig that has additional aeration tubes fitted with air stones installed under the separation chamber (145 mm long, 155 mm wide and 320 mm high) to generate air bubbles. The hybrid jig separation experiments were done by putting plastic samples and a solution (20 L) containing water, MIBC, and one of the wetting agents into the separation chamber (Table 2). Air bubbles were introduced underneath the screen simultaneously as water pulsation begun (Fig. 1(b)). Table 2 summarizes the conditions of hybrid jig separation experiments conducted in this study. The water pulsation was decided based on the research of Hori et. al., 2009 [4]. After each run, the products were divided from the top into six layers (Fig. 1(c)) and then collected using a vacuum sampling system. Different plastics in each layer were separated by hand-picking to measure the purity of products.

Table 2. Conditions of the hybrid jig separation tests.

	Experiment 1 HIPS/PPGF	Experiment 2 PVC/PA
Weight of sample [g]	250	
Particle size [mm]	+3.0 - 8.0	
Displacement [mm]	10	
Frequency of water pulsation [cycle/min]	30	
Separation time [min]	3	
Air rates [L/min]	1.5	
Concentration of stabilize bubble formation reagent [ppm or mg/L]	20	
Concentration of wetting agent [ppm or mg/L]	350 (NaLS, TA)	10 (AOT)

Results and discussion

Separation of PPGF and HIPS using bubble attachment probability

PPGF is typically used for drum-type washing machines while HIPS is a common type of plastic used in the manufacture of many home appliances because of its durability. Although the SG of PP is less than 1.0, addition of glass-fiber to create the stronger PPGF increased its SG to 1.04, which is equal to that of HIPS. Because both of these types of plastics are common components of large home appliances, they are mixed during recycling and should be separated to improve material recycling efficiency.

Preliminary experiments to separate PPGF and HIPS using the hybrid jig without any wetting agent showed that separation was impossible because of the indiscriminate attachment of bubbles on both types of plastics. Fig. 3(a) shows the hybrid jig experimental results for the PPGF-HIPS mixture (3.0 to 8.0 mm) in solutions containing 350 ppm of AOT, NaLS or TA. Purity in the top (1^{st}) and bottom (6^{th}) layers (Fig. 1(c)) with AOT (Fig. 3(a-1)) was lower than those with NaLS (Fig. 3(a-2)) and TA (Fig. 3(a-3)). With NaLS, the PPGF purity in the top layer and the HIPS purity in the bottom layer were 97% and 99%, respectively and with TA, the PPGF purity in the top layer and the HIPS purity in the bottom layer were 99% and 86%, respectively. These results suggest that separation of both plastics is possible, and the type of wetting agent and their concentrations are important parameters to improve the hybrid jig separation efficiency.

The effects of a wetting agent on particle-bubble interactions depend on tensile forces acting on the particle that lead to the development of an angle between the particle surface and the bubble surface [30]. At equilibrium

$$\gamma_{s/a} = \gamma_{s/w} + \gamma_{w/a} \cos\theta \quad (1)$$

where $\gamma_{s/a}$, $\gamma_{s/w}$, and $\gamma_{w/a}$ are the surface energies between solid and air, solid and water, and water and air, respectively, and θ is the contact angle (angles between particle surface (solid-liquid interface) and bubble surface (gas-liquid interface)) (Fig. 1). The attachment of bubbles on the surface of particles is strongly dependent on the work of adhesion ($W_{s/a}$), which is defined as the force required to break the particle-bubble interface and is equal to the work needed to separate the solid-air interface and produce separate solid-air, air-water and solid-water interfaces (Eq. 2) [30].

$$W_{s/a} = \gamma_{w/a} (1 - \cos\theta) \quad (2)$$

The work of adhesion is further influenced by the surface wettability of the particle, determined by the contact angle (θ), and the surface tension of solution. More hydrophobic (i.e., larger contact angle) particles have larger work of adhesion when in contact with bubbles. Similarly, solutions with higher surface tension ($\gamma_{w/a}$) increase the work of adhesion between particle and bubble. Both the surface tension and surface wettability of particles could be changed by adding surface acting agents called surfactants [30], so measurements of the contact angles of PPGF and HIPS as well as the surface tension of solutions containing each of the wetting agents were carried out.

Figs. 3(b) and 4 show the contact angles of PPGF and HIPS as well as the surface tension of solutions containing the wetting agents (i.e., AOT, NaLS or TA). The results showed that PPGF is more hydrophobic than HIPS regardless of the type and concentration of wetting agent (Fig. 3(b)).

Among the three wetting agents, only AOT lowered the surface tension of solution while the effects of NaLS or TA on it were negligible (Fig. 4). The lower surface tension of solution in the presence of AOT decreased the work of adhesion, resulting in fewer bubble attachments on the particle surface. In comparison, NaLS or TA had negligible effects on the surface tension but decreased the contact angle at higher concentrations (Fig. 4). These results suggest that the work of adhesion and bubble attachment probability decreased in the presence of either NaLS or TA likely because of the adsorption of these two wetting agents onto the surface of plastic particles that made them more hydrophilic. Similarly, the contact angles on both plastics decreased in the presence of AOT (Fig. 3(b-1)) but because this wetting agent also lowered the surface tension of solution (Fig. 4), the net effect was the limited bubble attachment that resulted in low separation efficiency even though both PPGF and HDPS became more hydrophilic (Fig. 3(a-1)). In the case of NaLS, the change in contact angle of HIPS was more dramatic than that of PPGF, which caused selective bubble attachment onto PPGF making it lighter than HIPS that facilitated better stratification during hybrid jig separation (Figs. 3(a-2) and 3(b-2)). In the case of TA, the contact angle of PPGF decreased with increasing TA concentration (Fig. 3(b-3)). However, the contact angle with TA was slightly higher than that with AOT, which likely caused selective bubble attachment on the PPGF surface. These results show that the control of surface characteristics by wetting agents could improve the separation efficiency of the hybrid jig. In the TA solution, similar results were obtained compared with that in NaLS even though the contact angles of HIPS and PPGF were similar. This phenomenon remains unclear because of the difficulty in quantifying the amounts of bubbles attached on plastic particles. Many previous papers reported the interaction between air bubbles and plastic particles during flotation [35, 36] however, the particle and water movement during hybrid jig separation and flotation are different. This means that a reliable measurement technique to quantify bubble attachment volume during hybrid jig separation (with water pulsation) should be developed.

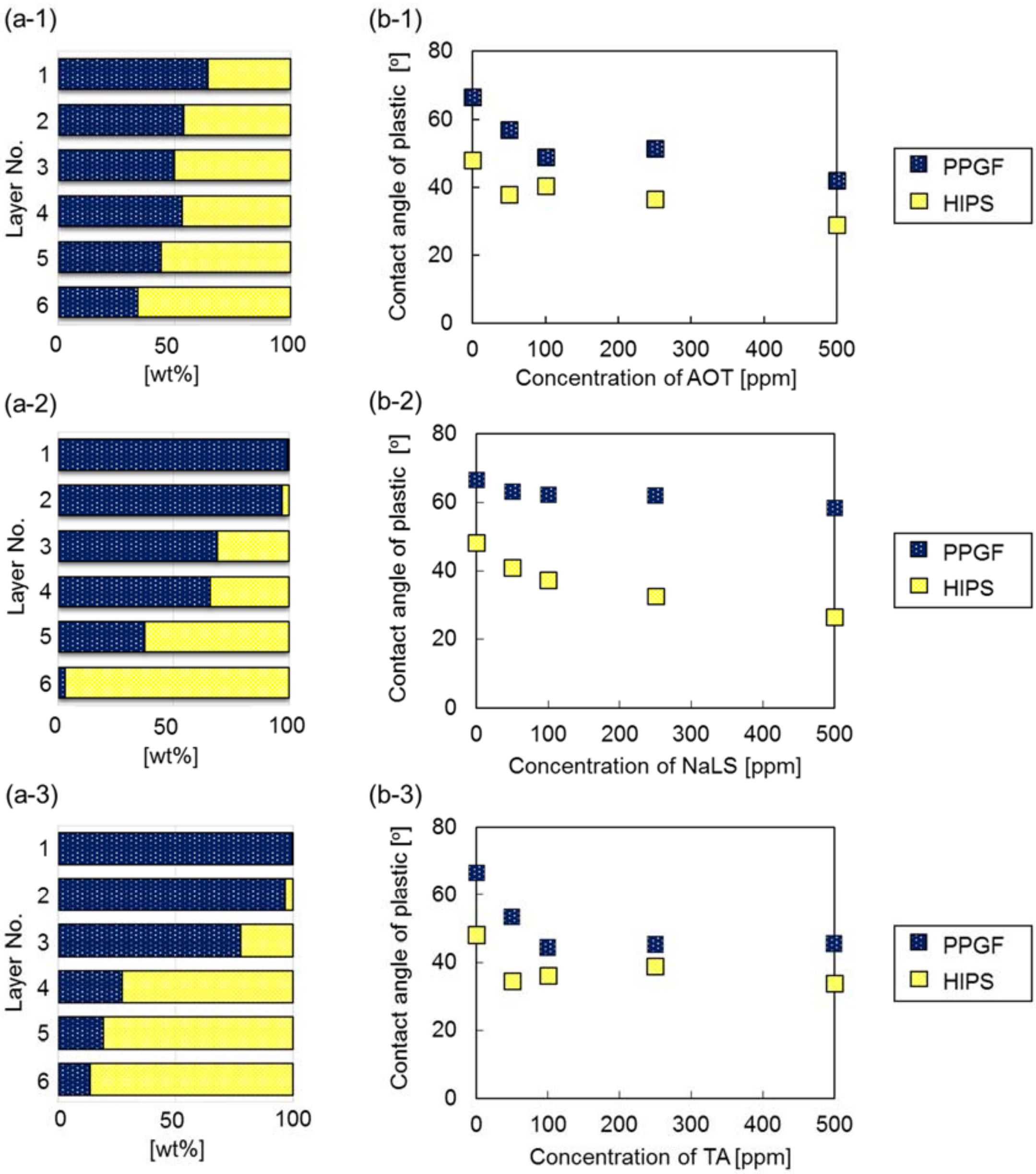

Fig. 3 (a) The proportion of PPGF and HIPS in each layer after hybrid jig separation tests with (a-1) AOT (350 ppm), (a-2) NaLS (350 ppm), and (a-3) TA (350 ppm) as wetting agents, and (b) contact angles of PPGF and HIPS in (b-1) AOT, (b-2) NaLS, and (b-3) TA solutions

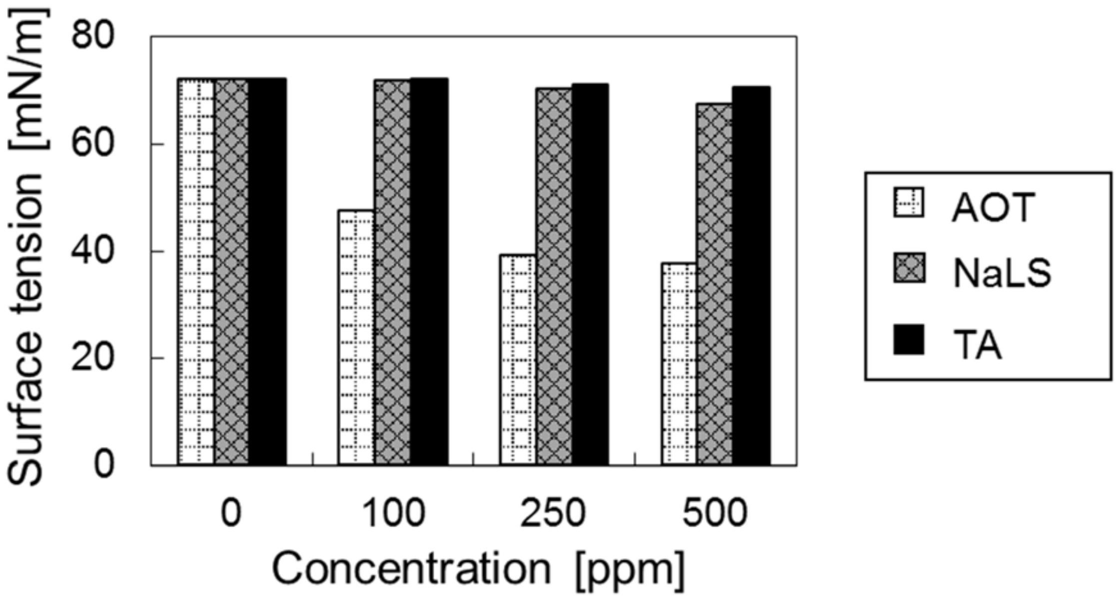

Fig. 4 Surface tension of solutions containing each of the wetting agents at various concentrations

Separation of PVC and PA using a two-step approach

PVC is a common type of plastic used for a variety of applications such as in building and construction, packaging, electrical and electronics, and automobile while PA (or nylon) is an engineering plastic used in clothing, electrical and electronic, and automobile [1]. These types of plastics can be found together in waste electrical and electronic equipment (WEEE) and automotive shredder residue (ASR) from end of life vehicle (ELV) and due to their similar SG (1.38 and 1.37), in this study hybrid jig was applied to separate them into individual components to improve material recycling efficiency.

The results from the previous section showed that wettability control by selective bubble attachment with the aid of wetting agents could improve the hybrid jig separation efficiency. In this section, separation of PVC and PA using a two-step approach was investigated. Hybrid jig tests of PVC (SG = 1.38) and PA (SG = 1.37) were carried out in water containing 10 ppm of AOT. The results showed that these two plastics could not be separated without AOT (i.e., separation efficiency was almost 0%) because of the indiscriminate attachment of bubbles to both types of plastics. Fig. 5(a-1) shows the result of hybrid jig separation with 10 ppm of AOT. The PVC purity in the top layer and the PA purity in the bottom layer were 84% and 98%, respectively. Separation of PVC and PA occurred because AOT lowered the contact angle of PA but not that of PVC as shown in Fig. 5(b). However, bubble attachment probability on PVC slightly decreased with AOT addition (Fig. 5(b)) due to decrease of surface tension as discussed earlier (Fig. 4), that likely caused imperfect separation (Fig. 5(a-1)).

To improve the separation efficiency, a two-step hybrid jig method (pre-wetting of PA) is proposed. In the first step, plastic mixture of PVC and PA are put in 10 ppm AOT solution for 3 min. Then, solution is discharged, and water is filled instead (second step) and hybrid jig separation in water is carried out. Fig. 5(a-2) shows that higher purities of PVC (94%) in the top layer and PA (99%) in the bottom layers were obtained compared with the single-step approach because higher surface tension of second step method than one-step method caused better bubble attachment on PVC (Fig. 5(a-1)). However, due to the replacement of water in second step may cause some effect thus, effects of time after water replacement was checked. The results showed that hybrid jig separation just after water replacement was effective (Fig. 5(a-2)) however, several tens of minutes after water replacement showed worse hybrid jig separation efficiency because bubble attachment to the PVC became difficult again. This lower bubble attachment probability of PVC may cause by the decrease of surface tension due to the detachment of AOT from PA surface. To understand the attachment and detachment behaviors of AOT on PA, the adsorption and desorption experiments were carried out. The adsorption of AOT on PA was confirmed (Fig. 6(a)) and desorption of AOT from PA with time (Fig. 6(b)) showed that AOT was readily desorbed causing the surface tension of solution to decrease with time (Fig. 6(b)). In other words, the separation results become bad with time because the wettability of PVC was enhanced due to the decrease in surface tension of solution caused by the desorption AOT adsorbed on PA during the pre-wetting stage. This means that although pre-wetting was effective, the adsorption-desorption behavior of wetting agent could strongly affect the separation efficiency, so precise control of the wettability of samples is required.

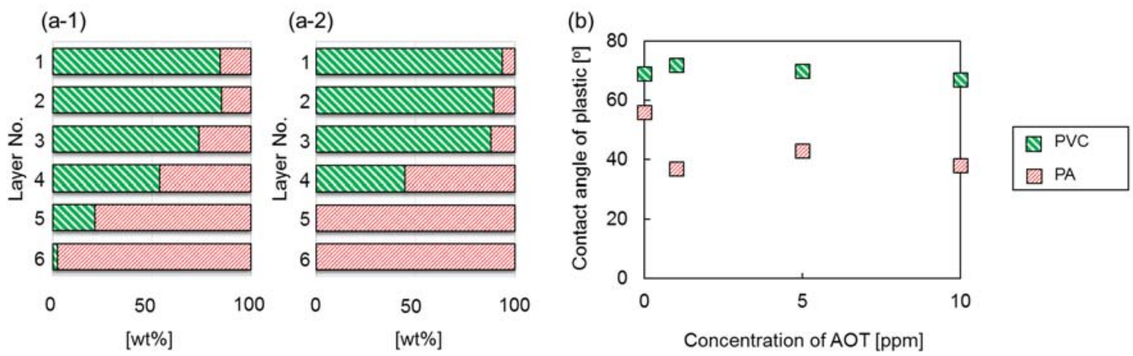

Fig. 5 (a) The proportion of PVC and PA in each layer after hybrid jig separation tests (a-1) in AOT (10 ppm) solution (one-step method) and (a-2) after solution replacement with water (two-step approach), and (b) contact angles of PVC and PA in the AOT solutions

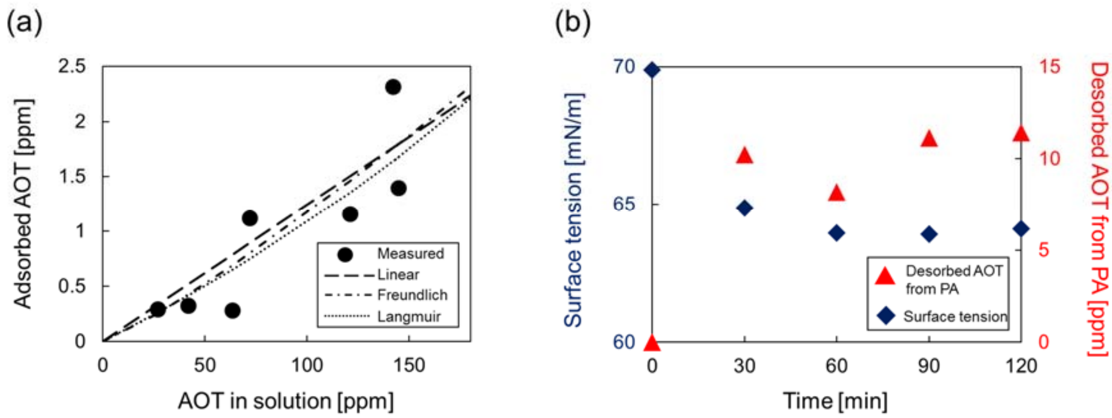

Fig. 6 (a) Adsorption characteristics of AOT onto PA with linear, Freundlich and Langmuir isotherms, and (b) desorbed AOT concentration from PA with time and the corresponding changes in the surface tension of the solution

Conclusions

This paper evaluated the improvement of hybrid jig separation of mixed-plastic wastes using wetting agents and the findings of this study are summarized as follows:

- Wetting agents could change the surface tension of solutions by changing the water-air surface properties and the surface wettability (contact angle) of plastics by surface adsorption, both of which lowered the bubble attachment probability.
- For the separation of PPGF and HIPS, surface modification using wetting agents (i.e., AOT, NaLS or TA) could improve the separation efficiency.
- In the case of PPGF/HIPS with AOT, the changes in surface tension and the decrease of contact angles of both plastics caused lower bubble attachment probability, so selective attachment of bubbles was limited that caused lower separation efficiency.
- In the case of PPGF/HIPS with NaLS, the changes in surface tension were negligible and the contact angle on HIPS decreased while that on PPGF was unchanged, so selective attachment of bubbles on PPGF occurred and higher separation efficiency was obtained.
- In the case of PVC/PA with AOT, the surface tension decreased and the contact angle on PA decreased while that of PVC did not change, which caused selective attachment of bubbles on PVC.
- A two-step approach could increase the surface tension, increase the bubble attachment probability on PVC, and dramatically improved the hybrid jig separation efficiency.

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